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“THE ORDER OF CRYSTALLIZATION IN IGNEOUS ROCKS”

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In a recent number of this *Journal* appeared an article entitled “The Order of Crystallization in Igneous Rocks.”¹ A series of conclusions far-reaching in their effect on petrography are arrived at by the author, which are certainly worthy of further comment and discussion. By building up on certain fundamental assumptions, the author is enabled to give a series of diagrams showing for each of the important rock groups both the order of beginning and the order of cessation of crystallization. A further discussion of the assumptions seems, on account of the importance of the conclusions arrived at, very desirable.

In considering such a subject as the crystallization of a magma, we must first of all remember that we have to deal with a definite chemical system whose behavior is rigidly governed by the laws of mass action and the phase rule. The laws of physical chemistry should throw some light on this problem, and it is the purpose of the author to advance some ideas which may affect the conclusions arrived at in the above article.

Can we regard a granite and a rhyolite as representing the same chemical system? That is, may we in the full sense of the term regard a rhyolite as the quenched portion of the chemical system, which, if undisturbed, would result in a granite?

Geologists generally agree that the same magma, depending on certain variable conditions, such as temperature, pressure, and rate of cooling, may yield either a granite or a rhyolite, but probably none will maintain that these variable factors, and especially temperature and pressure, will have an effect only on the physical condition, but not also on the chemical condition of the magma. A granite consists not only of a solution of the various oxides in

¹ N. L. Bowen, *Journal of Geology*, XX, 455.

each other which subsequently enter into the formation of the different minerals in the rock, but there also enter in a series of volatile components such as water, boron, fluorine, chlorine, etc.—the mineralizers. How are these held in the magma? In all probability they are in certain molecular combination with some of the other oxides present. Under the conditions of great pressure and in the presence of mineralizers, a granite begins and completes crystallization. The effect of these volatile components is considered so important that the solidification temperatures of certain granites is placed as low as 200° – 350° C.¹ These mineralizers are the powerful solvents, they represent to a large extent the “mother liquor” from which the granite crystallized.

Turn now to the case of a rhyolite. The variable factors are quite different. Pressure is far less, the rate of cooling is far more rapid. What will be the effect of such a change? In physical chemistry we recognize what is known as van't Hoff's law, which deals with the fact that displacements of equilibrium within a solution are effected by changes in temperature and pressure. Crystallization in a magma can be brought about in one case at a higher and in another case at a lower temperature, depending on the pressure. An increase of pressure will induce crystallization at a higher temperature. By a change of pressure equilibrium will be destroyed within the magma, and reactions taking place will go farther in one direction, thus altering the relative proportions of the various compounds present. As a result we may obtain in the rock an association of minerals quite different from what it would have been under a different pressure.² As a further result of decrease of pressure, the volatile constituents would escape, causing additional disturbance in the equilibrium and rearrangement in the molecular grouping, which will further influence the final crystallization product. Thus the law of mass action and van't Hoff's law seem to prove that granite and rhyolite cannot safely be regarded as having the same order of crystallization nor the same mineral make-up. The conception of mineralizers strengthens this view.

¹ Harker, *The Natural History of Igneous Rocks*, p. 189.

² Cf. C. N. Fenner, *American Journal of Science*, XXIX (March, 1910), 217.

If a solution crystallizes, is a recurrence of the first component to solidify possible, after complete crystallization of the other dissolved substances? We must concede as true that a magma represents a solution at high temperature, and that its crystallization must be analogous to the crystallization of any solution. Chemical precipitation due to chemical reaction caused by concentration, or by the elimination of some dissolved substance, may take place, but on the whole the analogy to a freezing salt solution should be striking. This has been pointed out repeatedly by various authors.

For comparison, let us recall Usiglio's classical experiments on the evaporation of sea-water. In this case we have a complex solution of such compounds as Fe_2O_3 , CaCO_3 , CaSO_4 , NaCl , KCl , MgSO_4 , MgCl_2 , etc. Pressure and temperature were kept constant, but the degree of concentration was varied. The following order of crystallization was obtained: Fe_2O_3 , CaCO_3 , $\text{CaSO}_4 + 2 \text{H}_2\text{O}$, NaCl , MgSO_4 , MgCl_2 , NaBr , KCl . There was an overlapping of the periods of crystallization, but in no case was there a recurrence of the crystallization of any member after a later product had formed (see fig. 1). When a salt solution freezes, ice crystallizes out, and the solution becomes more and

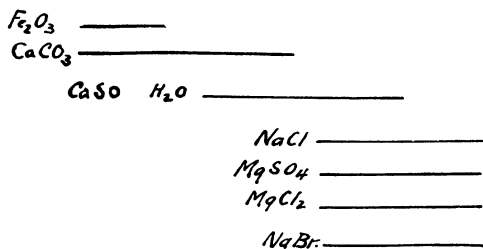


FIG. 1.—Order of crystallization upon partial evaporation of sea-water.

more concentrated until saturation is reached (23.6 per cent NaCl); when at a temperature of -22°C . both crystallize together, forming a cryohydrate. Salt does not crystallize out to be followed later by ice. Other examples of crystallization of salt solutions might be cited, but the above are sufficiently familiar and characteristic. In all cases (see Figs. 2, 3), however, no overlap of the first crystals formed can occur with regard to later products, as far as order of cessation of crystallization is concerned. In the majority of cases the end product should be a cryohydrate (eutectic).

For a different case let us assume we have three metals fused into an alloy. Provided they are completely soluble in each other, the eutectic ratio is the controlling factor, the metal in greatest excess crystallizing first, until the second metal reaches its saturation point, when the two come down together, until finally the ternary eutectic point is reached and crystallization of all goes on simultaneously. In case the metals interact, chemical compounds may be formed and complicate the system. Thus in a combination

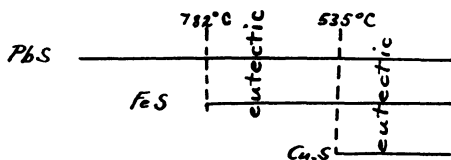


FIG. 2.—Order of crystallization in alloy of PbS=70 per cent, FeS=20 per cent, Cu_2S =10 per cent.

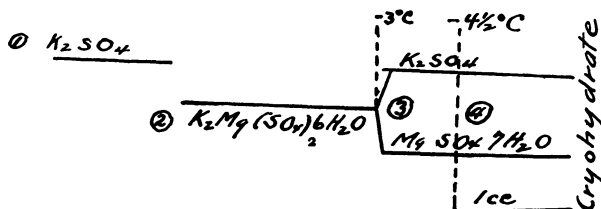


FIG. 3.—Crystallization of K and Mg sulphate solution, when K_2SO_4 is in excess and saturates solution between 92° and -3° C. Schönite (2) is stable only between 92° and -3° C. See van't Hoff's *Theoretical and Physical Chemistry*, p. 86.

of bismuth, lead, and tin, the order of beginning of crystallization will depend on relative masses and will end with a ternary eutectic. In the case of a copper, tin, and antimony mixture, we get chemical reactions during cooling which may result in the formation of three pure metals and seven chemical compounds. There is no recurrence of the substance first crystallized in either case. If substances are capable of forming solid solutions they do not affect the above in any way.

For sake of simplicity let us assume we have a magma of SiO_2 , KAlSi_3O_8 , and $\text{NaAlSi}_3\text{O}_8$, an example worked out by Harker.¹

¹ Harker, *Natural History of Igneous Rocks*, p. 251.

The order of crystallization will depend on relative masses, but will in every case end with a ternary eutectic. We all concede that the rate of growth of different crystals varies, but it does not seem probable that in the same solution one substance, such as quartz, begins solidifying before a second, say orthoclase, and also is still crystallizing after the latter is completely removed. Such overlapping would seem to be an unusual exception to the general rule, of which no example has been found. Considering this point, may we not assume that order of cessation of crystallization gives a definite clue to the order of beginning of crystallization?

SUMMARY

With a view of testing the conclusions as to order of crystallization, the author advances the following points:

1. We cannot assume that the effects of pressure and of crystallization temperatures may be considered negligible in the case of rhyolite and granite (effusive and deep-seated rocks). Therefore it is doubtful that we can assume the same order of crystallization or the same end-products.

2. We must recognize the importance of the mineralizers, not only as affecting the physical condition of the magma, but also its molecular arrangement, and consequently its manner and possibly, order of crystallization.

3. Comparing the order of crystallization in solutions and alloys, we find that the order of cessation of crystallization gives a definite clue as to the order of beginning of crystallization, consequently overlapping both of the beginning and the cessation of crystallization of any substance by another seems improbable.

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